# Accelerated and natural carbonation of concrete with high volumes of fly ash: chemical, mineralogical and microstructural effects

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## Introduction

Fly ash (FA) already has a long history of being used as partial replacement of ordinary Portland cement (OPC) in concrete. When focusing on high-volume fly ash (HVFA) concrete, special attention should go to environments subject to atmospheric $CO_2$ ingress and carbonation-induced steel corrosion. In OPC concrete, $CO_2$ dissolves in the pore solution to form carbonic acid which reacts with $Ca(OH)_2$ (CH) and calcium silicate hydrates (C–S–H) in the cement paste, forming mainly $CaCO_3$. Although the latter results in a more dense microstructure, the related drop in pore fluid alkalinity can disrupt the protective passivation layer on embedded steel and cause active corrosion [1]. In HVFA concrete, the pozzolanic hydration reaction of FA also consumes CH [2]. As a result, less CH is available and the carbonation front moves inwards faster. Moreover, the lower CH availability will result in more C–S–H carbonation [3]. In contrast with CH carbonation, C–S–H carbonation coarsens the pore structure. Also note that CH carbonation precipitates mainly well-crystallized calcite, while its amorphous and metastable polymorphs (vaterite and aragonite) are more likely the result of C–S–H carbonation [4]. The higher susceptibility of FA binders to carbonation is usually concluded from accelerated tests at high $CO_2$ levels. The applied $CO_2$ concentration ranges from 1% to 100% [5]. For OPC binder systems it has already been demonstrated that accelerated test conditions could alter the carbonation process, yet for HVFA binder systems this was studied less. In this research, the chemical, mineralogical and microstructural effects of accelerated carbonation at 10% and 1% $CO_2$ as opposed to natural carbonation at 0.03% $CO_2$ have been investigated for pastes with a 50% and 40%+10% replacement level of the OPC by FA and fly ash + silica fume (FA+SF), respectively. CH to C–S–H carbonation ratios were quantified from thermogravimetric analyses (TGA). Related mineralogical phases were identified with X-ray Diffraction (XRD) and changes in pore size distribution and porosity were determined via Mercury Intrusion Porosimetry (MIP).

## Conclusions

When field carbonation rates are estimated from an accelerated carbonation experiment at 1% $CO_2$ cf. [6], these rates exceed by far the ones converted from carbonation expriments at 10% $CO_2$ for OPC, HVFA and FA+SF concrete. The underestimation inherent to carbonation testing at 10% $CO_2$ cannot simply be explained by an important reduction in porosity and densification of the pore structure as assessed by means of MIP. On the other hand, TGA and XRD analyses indicate that the carbonation mechanisms during exposure to 1% and 10% $CO_2$ are similar. More important differences exist between natural carbonation at 0.03% $CO_2$ and slightly accelerated carbonation at 1% $CO_2$. More research is needed to come up with a more accurate formula that adequately converts an accelerated into a field carbonation rate.

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